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Glass transition in mixed network former glasses: Insights from calorimetric measurements

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Modifier-free mixed network former glasses are technically important due to their special properties, e.g., low thermal expansion coefficient, low Young's modulus, and high strain points (≥650 ℃).[1)] In addition, many mixed network former glasses have high liquidus viscosities (>3×10^5 poise), which is a requirement for the fusion sheet manufacturing process.[2)] Some previous studies have shown the evolution of structural units with composition in ternary network former glasses.[1),2)] Moreover, it has been reported that there is a preference for Al-P instead of B-P association in quaternary network former glasses.[3)] In order to explore the change of the degree of connectivity in mixed network formers with composition, it is highly interesting to study the composition dependent glass transition temperature (T_g) of mixed network former glass. Now the questions arise: For this type of glass, how do the thermodynamic properties evolve with composition, such as T_g and C_p jump (ΔC_p) during glass transition? By performing calorimetric measurements, can we gain an insight into the degree of distribution of different network formers? In this work, we explore the thermodynamic properties of B_2O_3-SiO_2-P_2O_5-Al_2O_3 mixed network former glasses with a substitution of B_2O_3 for SiO_2 using differential scanning calorimetry (DSC). Figure 1 shows that there are multiple crystallization peaks and two glass transition regions for each sample, and the addition of B_2O_3 causes the number of crystallization peaks increasing and the crystallization temperature decreasing. It has been recently reported the existence of three T_gs in an ionomer glass.[4)] Although our studied compositions are far away from the immiscible region in the B_2O_3-P_2O_5-SiO_2 phase diagram if Al_2O_3 (4 mol% in all studied glasses) is considered to be replaced by B_2O_3, two T_gs exist in all studied glasses, implying the presence of two non-crystalline phases[4)]. As shown in Figure 2, both of the two T_gs decrease linearly with addition of B_2O_3 content, suggesting that the network would be depolymerized with further addition of B_2O_3. Moreover, T_{g1} decreases much faster than T_{g2}. The different changing rate of two T_gs may lead to T_g merging in a critical composition if we extrapolate the linear fitting lines as shown in Figure 2. As a measure of indirect thermodynamic fragility, the ΔC_p during the glass transition is calculated. It increases about 20% when B_2O_3 content increasing from 16 mol% to 27 mol%, indicating that the mixed network former liquid becomes more fragile with the increase of B_2O_3 content.

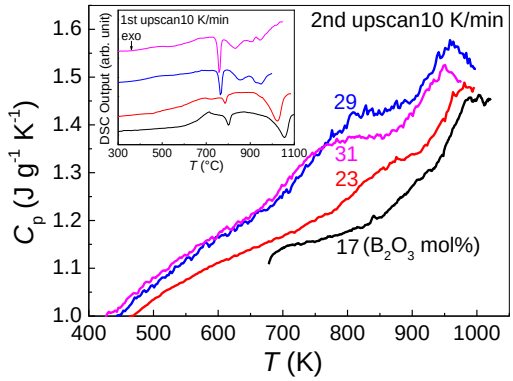

Figure1. The C_p curves for the 2nd upscan at a heating rate of 10 K/min subsequent to a cooling procedure at the same rate. Inset: DSC output for the 1st upscan at a heating rate of 10 K/min.

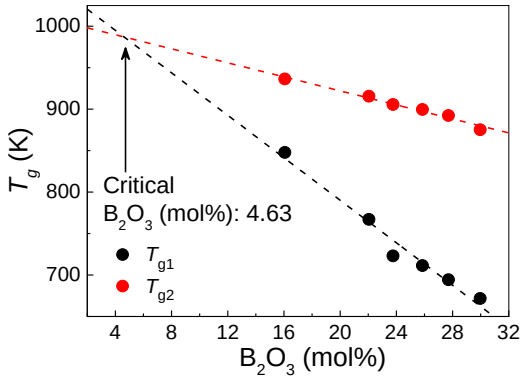

Figure 2. The evolution of T_g with composition. The dash lines are linear fitting.

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